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Title: **PERFORMANCE TESTING OF A LARGE VOLUME CALORIMETER**

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Performance Testing of a Large Volume Calorimeter

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Abstract

Calorimetry is used as a nondestructive assay technique for determining the power output of heat-producing nuclear materials. Calorimetric assay of plutonium-bearing and tritium items routinely obtains the highest precision and accuracy of all nondestructive assay (NDA) techniques, and the power calibration can be traceable to National Institute of Standards and Technology through certified electrical standards. Because the heat-measurement result is completely independent of material and matrix type, it can be reliably used on any material form or item matrix. The calorimetry measurement is combined with isotopic composition information to determine the correct plutonium content of an item. When an item is unsuitable for neutron or gamma-ray NDA, calorimetric assay is used. Currently, the largest calorimeter capable of measuring plutonium-bearing or tritium items is 36 cm in diameter and 61 cm long.

Fabrication of a high-sensitivity large volume calorimeter (LVC) capable of measuring tritium and plutonium-bearing items in 208-l (55-gal) shipping or storage containers has provided a reliable NDA method to measure many difficult to measure forms of plutonium and tritium more accurately. This large calorimeter can also be used to make secondary working standards from process material for the calibration of faster NDA assay techniques. The footprint of the calorimeter is 104 cm wide by 157 cm deep and 196 cm high in the closed position. The space for a standard electronics rack is also necessary for the operation of the calorimeter. The maximum item size that can be measured in the LVC is 62 cm in diameter and 100 cm long.

The extensive use of heat-flow calorimeters for safeguards-related measurements at DOE facilities makes it important to extend the capability of calorimetric assay of plutonium and tritium items to larger container sizes.Measurement times, precision, measurement threshold, and position sensitivity of the instrument will be discussed.

Introduction

Calorimetry is used as a nondestructive assay technique for determining the power output of heat-producing nuclear materials. Calorimetric assay of plutonium-bearing and tritium items routinely obtains the highest precision and accuracy of all nondestructive assay (NDA) techniques, and the power calibration can be traceable to National Institute of Standards and Technology through certified electrical standards [1]. Because the heat-measurement result is completely independent of material and matrix type, it can be reliably used on any material form or item matrix. The single assumption is there are no endothermic or exothermic chemical reactions occurring in the item. The calorimetry measurement is combined with isotopic composition information to accurately quantify the plutonium content of an item. When an item is unsuitable for neutron or gamma-ray mass measurement (i.e. TGS, SGS) NDA or destructive analysis, calorimetric assay is normally used when possible. Currently, the largest calorimeter capable of measuring plutonium-bearing or tritium items is 36 cm in diameter and 61 cm long. Larger calorimeters for the measurement of tritium [2,3] and animals [4] have been fabricated previously.

A calorimeter capable of measuring the power output from 208 liter (55-gal) drums was designed and fabricated at Los Alamos National Laboratory (LANL). The fabrication of this high-sensitivity Large Volume Calorimeter (LVC) capable of measuring tritium and plutonium-bearing items in 208-l shipping or storage containers has provided a reliable NDA method to measure many difficult to measure forms of plutonium and tritium more accurately, in the Department of Energy (DOE) complex. This large calorimeter can also be used to make secondary working standards from process material or waste material categories for the calibration of faster NDA assay techniques.

Calorimeter Design

The LVC uses thermopile heat-flow sensors as a replacement for the Wheatstone bridge sensors that are used in a majority of the radiometric calorimeters in the United States. The thermopile heat-flow sensors were supplied by International Thermal Instrument Company [5]. The footprint of the calorimeter is 104 cm wide by 157 cm deep and 196 cm high in the closed position. The space for a standard electronics rack is also necessary for the operation of the calorimeter. A standard 208-l drum with a 60 cm diameter and retaining ring with bolt and up to 100 cm long can be measured in the LVC. With special positioning considerations cylindrical items up to 66 cm diameter could be measured. Data acquisition and instrument control are managed with the LANL-developed MultiCal program [6].

The loading/unloading of drums into and out of the LVC is done using a custom manufactured Versa-lift [7] drum handler. The 208-l drums are lifted and placed onto the LVC pedestal using the drum handler. The LVC pedestal is exposed by lifting the entire LVC shell and sensors. A pair of photographs is presented in Figure 1 of a 208-l drum being loaded into the LVC. The LVC pedestal is a circular insulating plug of extruded polystyrene that prevents item heat from being lost out the bottom end of the calorimeter. Interleaved between the pedestal insulation are two 1 mm thick stainless steel sheets used as heat shunts and two 1 mm thick silicone rubber encapsulated wire surface heaters. The outer heater is maintained at a constant temperature of 32 °C and the inner heater is maintained at a constant temperature of 36 °C. An aluminum plate with a counter sunk center is placed on top of the pedestal to provide additional heat shunting and centering of the item drums on the pedestal.

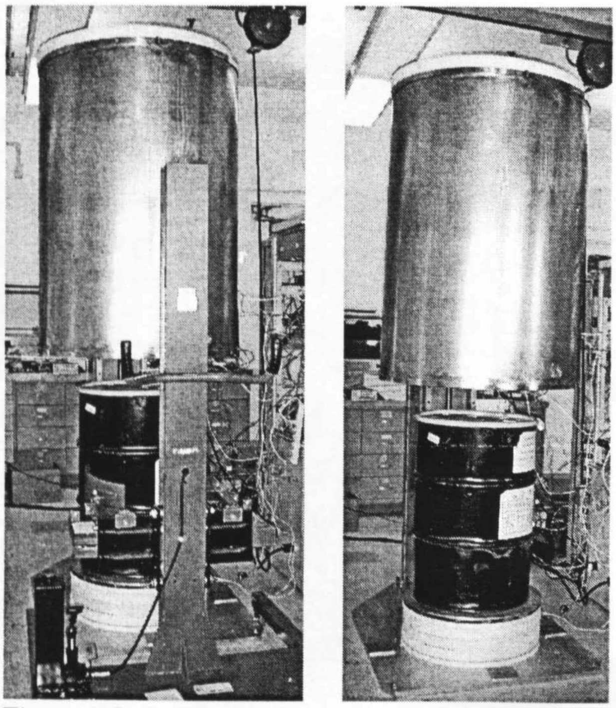

Figure 1: Left: Photograph of a 208-l drum being loaded into the LVC using a custom built drum handler. Right: Photograph of LVC with calorimeter in up position after 208-l drum has been loaded. The circular insulation below the drum is the LVC pedestal.

The calorimeter consists of three concentric cylinders closed on the top and open on the bottom for the insertion of the 208-l drums and pedestal. Figure 2 presents a top view of the LVC with the lids off the two outer cylinders. The outer most heater blanket is also labeled in Figure 2. The "outer can" is stainless steel with and OD of 84 cm and is designed to provide mechanical support for the instrument when lifted. The "mid can" is fabricated from rolled aluminum with an OD of 85 cm. The "sensor can" is also rolled aluminum with and OD of 69 cm and a welded lid.

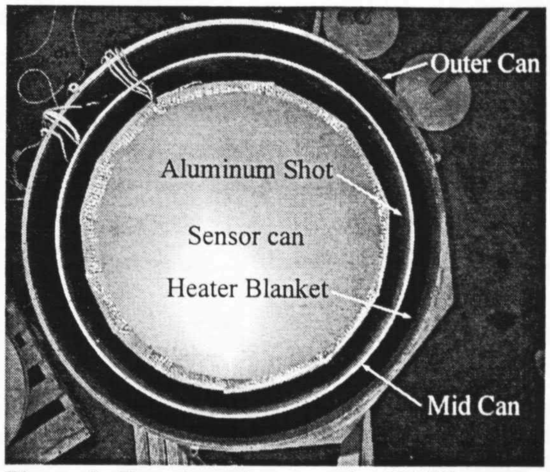

Figure 2: Top view photograph of LVC cylinders with lids off.

The LVC uses 2 conductive temperature zones heated by silicone rubber encapsulated wire surface heaters to provide a constant reference temperature to the cold side of the thermopile heat-flow sensors. The inner heater zone, at the outer surface of the sensor can is controlled to the same temperature as the inner pedestal heater, 36 °C. There is also an inner lid and annulus heater all maintained at a temperature of 36 °C. Temperature control is achieved via servo controlled feedback loops for each heater. There are a total of 8 servo controlled heaters: inner cylinder, lid, annulus, and pedestal and outer cylinder, lid, annulus, and pedestal. The temperature feedback signal is obtained from each heater via a four wire resistance readout of a thermistor. The outer heater zone is maintained at a temperature of 32 °C. The outer heaters are on the inner surface of the "outer can" as labeled in Figure 2. Position and control temperature are the only differences between the inner and outer heater zones. Insulation and thermal mass are used in conjunction with the feedback loops to maintain stable reference temperatures and maintain a temperature differential between the inner heater zone, the outer heater zone, and room temperature. The LVC does not use any water or other significant neutron moderating or reflecting materials for temperature control. The LVC does not have the ability to actively cool so the instrument must be run in rooms colder than 28 °C.

In order to maintain a relatively small overall size the LVC does not use any compensating chamber, to reduce thermal noise in the reference temperature. Drift of the reference temperature is the largest source of noise in the system.

Presented in Figure 3 is the sensor can with thermopile sensor bars attached, before application of the inner silicone rubber encapsulated wire surface heater. There are 21 sensor bars around the circumference of the sensor can. Each sensor bar is 5 cm wide and mounted 10.4 cm on center from the adjacent sensor bar. Half the length of each sensor bar is active sensor alternating between dead areas (due to electrical leads, mechanical support, and mounting) and thermopile junctions every 5 cm. Every 5cm by 5cm active sensor area contains 240 bismuth-telluride thermocouple junctions. The thermal conductivity of the sensors in the direction of heat flow is 1.9 Btu/hr. ft. °F.

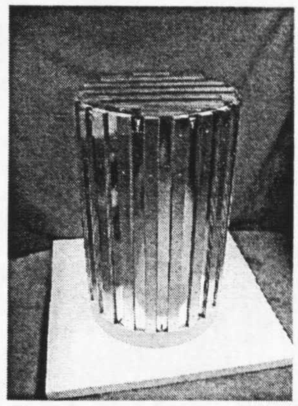

Figure 3: Photograph of LVC "sensor can" with thermopile sensor bars attached.

Performance

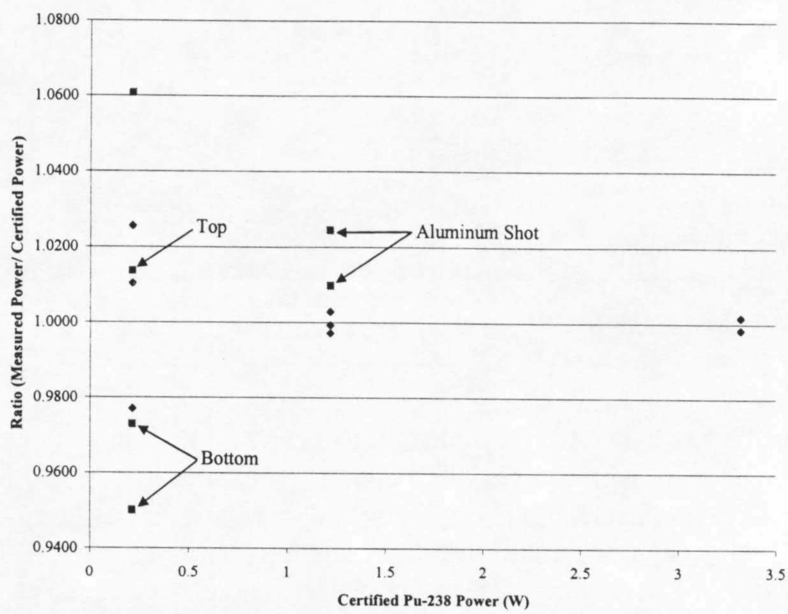

Figure 4: LVC bias is plotted as the ratio of the measured Pu-238 heat standard power to the certified power value as a function of power.

The measured LVC bias is presented in Figure 4 as a ratio of the measured source power to the certified source power as a function of certified source power. The diamonds represent data that was used to calibrate the instrument to heat. The squares are additional measurement data not used in the calibration. The heat sources used were National Institute of Standards and Technology (NIST) traceable Pu-238 heat sources. Several measurement conditions were included in this data set. The standard measurement configuration for the data set in Figure 4 was a Pu-238 heat source placed in the center of a 208-l drum filled with crumpled aluminum foil. The total drum weight for the standard configuration was less than 15 kg. Two of the measurements represented by squares labeled "Aluminum Shot" in Figure 4 were taken with the source in the top third of an aluminum shot filled 208-l drum. This drum contained over 225 kg of aluminum shot. The time to equilibrium was significantly increased but the equilibrium values were the same as the standard drum configuration. The LVC power determination shows no matrix dependence as long as the item is allowed to reach equilibrium. The time to equilibrium increase is due to the large heat capacity of the aluminum shot filled drum.

Two position sensitivity configurations were measured at a power of 0.214 W. The first source position was on top of a 5 cm thick piece of insulation on top of a 208-l drum filled with crumpled aluminum foil, labeled "Top" in Figure 4. This configuration was used to keep the source heat at the top of the calorimeter. There was no measurable position sensitivity in the top configuration at a power of 0.214 W. The second position sensitivity test was performed with a 0.214 W source place on the center of the insulation pedestal that plugs the bottom of the calorimeter during measurements, labeled "Bottom" in Figure 4. Nothing else was in the calorimeter. The source in the bottom position sensitivity case is more extreme that any 208-l drum could be, since a drum would conduct heat through the measurement area of the calorimeter better than the still air. The two measurements made in the bottom sensitivity configuration are the two lowest diamonds at a power of 0.214 W. The number of points at 0.214 W and the overall spread of the data at all powers suggests that the two "low" points at 0.214 W are within statistical fluctuations.

The average bias for all of the measurements in Figure 4 is 0.3%. As expected the calorimeter measurements are essentially bias free, this is including the position sensitivity data taken at a power of 0.214 W and matrices with extremely different thermal properties.

Data taken below the lowest calibration power of 214 mW is presented in Figure 5 along with the data taken at 214 mW and 1.2 W. The lowest power measured was 28.7 mW (11.5 g low burnup plutonium equivalent). Measurement uncertainty increases with decreasing power and is largest at 28.7 mW. The low average bias and large standard deviations of the three lowest power measurements presented in Figure 5 are dominated by a single measurement that was biased much lower than the other replicate measurements. These single low measurements at each power were likely measurements that did not reach full equilibrium. If an item does not reach full equilibrium during the measurement time the measurement results will be biased low. The relative contribution of early equilibrium detection is largest at the lowest powers. The identification of early equilibrium is most likely to occur at the lowest powers due to the small signal making it difficult to determine if the calorimeter has reached equilibration. Precision on small power items may be improved by measuring the item longer than what would normally be judged equilibrium.

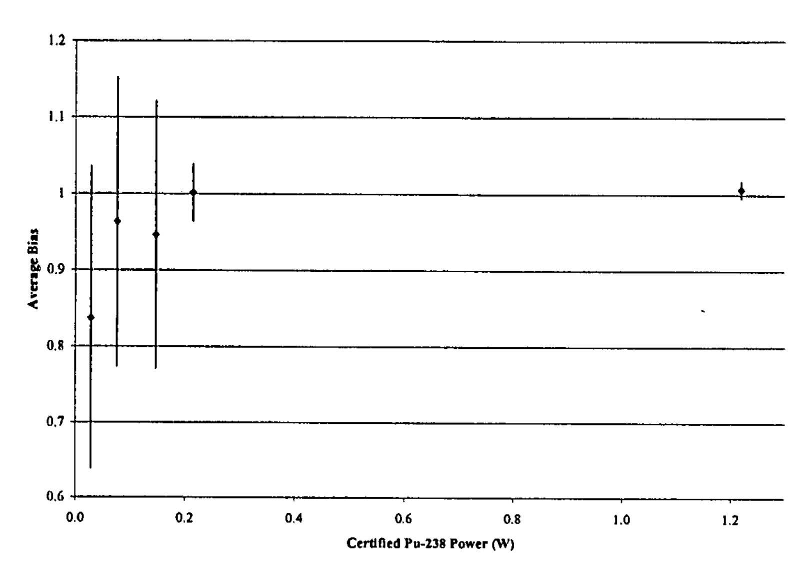

Figure 5: LVC average bias and one sigma uncertainty as a function of power.

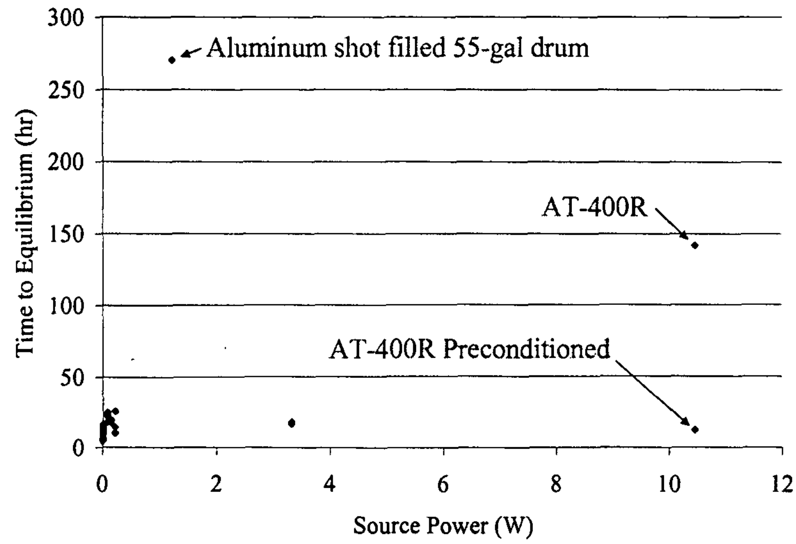

Figure 6: LVC equilibrium times for different powers, matrices, and starting temperatures. The average time to equilibrium for all measurements taking less than 30 hours is 14±5 hr.

The time it takes for the calorimeter to reach equilibrium is always an important parameter since calorimeter measurement time is usually greater than for other NDA techniques. Equilibrium time becomes a more important factor as the size and mass of the items being measured increases. The time necessary for a calorimeter to reach thermal equilibrium during the assay of an item is dependent on a number of factors such as: initial temperature of the item relative to the final equilibrium temperature of the item in the calorimeter (sample preconditioning can reduce measurement time by reducing this difference), type of heat-flow calorimeter used (passive or active), calorimeter size and thermal properties (thermal conductivity and total heat capacity) of the fabrication materials, thermal properties of the item and item packaging (usually more important than calorimeter properties), size and weight of the item and the calorimeter, use of an equilibrium prediction algorithm, and required assay accuracy [8] . Figure 6 presents equilibrium time data showing the variability of the time to equilibrium depending on measurement conditions.

The longest measurement time in Figure 6 is for a 1.25 W source in a aluminum shot filled 208-l drum. The long measurement time is due to the large heat capacity of the heavy drum. The two AT-400R type storage container points in Figure 6 give a measure of how much measurement time can be reduced by preconditioning the item before measurement. All of the other data points presented in Figure 6 are with the heat source in a foil filled 208-l drum (35 points), the source on the pedestal (1 point), or with the source on top of insulation at the top of 208-l drum (1 point).

The LVC equilibrium times less than 30 hours from Figure 6 are plotted as a function of starting room temperature in Figure 7. The time to equilibrium increases as the room temperature when the measurement was started decreases. A linear least squares fit is plotted in Figure 7 to demonstrate the trend in the data. The inverse proportionality between measurement time and starting room temperature is due to the final equilibrium temperature of the item in the calorimeter always being higher than the starting room temperature. The time to equilibrium did not show any correlations with source power or measurement date.

Conclusions

The LVC has dramatically increased the item size that can be measured using calorimetry to 208-l drums. The LVC is the first calorimeter of its kind capable of making matrix-independent high-precision power measurements on 208 liter drums. The LVC power measurements are independent of the position of the heat generating source within the drum. The LVC can be used to nondestructively assay difficult to measure forms of plutonium and tritium more accurately, make secondary working standards from process material or waste material categories for the calibration of faster NDA assay techniques, or make routine measurements on large quantity items with low measurement uncertainty. The power calibration is traceable to NIST through certified electrical standards.

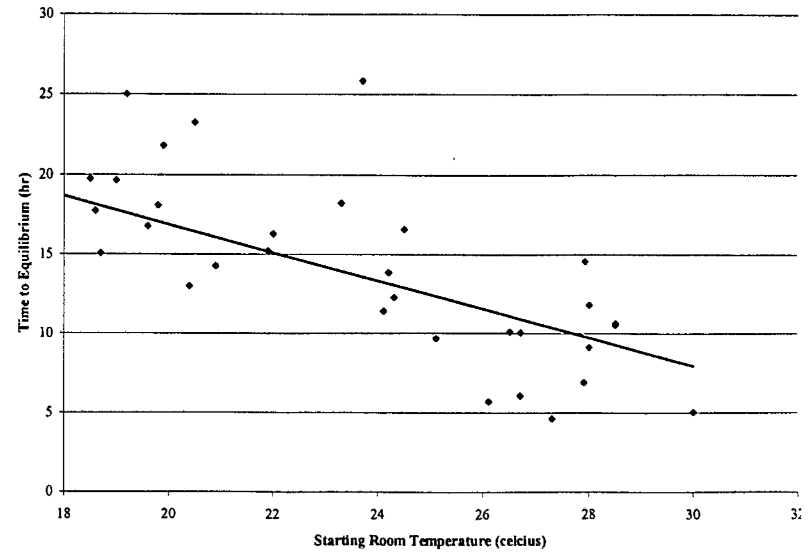

Figure 7: LVC equilibrium times as a function of room temperature when the measurement was started. The straight line is a least squares linear fit to the data used to guide the eye.

The footprint of the calorimeter is relatively small, 104 cm wide by 157 cm deep and 196 cm high, by not using water or a compensating chamber. Data acquisition and instrument control are managed with the user friendly LANL-developed MultiCal program that is currently in use with most calorimeters in DOE facilities.

The LVC has a high sensitivity, 119.61 mV/W, and relatively low measurement threshold for its size and is capable of measuring a wide range of plutonium or tritium quantities in all matrices that are thermally inert. Average measurement times for a low heat capacity 208-l drum (i.e. crumpled aluminum foil or air) is 14±5 hr. Equilibrium prediction can be used on this instrument to decrease measurement time. Equilibrium prediction would be most useful on items with a large heat capacity and high thermal power. Measurement threshold is less than 12 g low burnup plutonium. Measurement precision is 3.6 mW (0.3%) at a source power of 1.2 W. This measurement uncertainty is equivalent to 1.4 grams of low burnup plutonium.

Acknowledgments

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